# Economical Production of Pu-238: Feasibility Study

NASA NIAC Phase I

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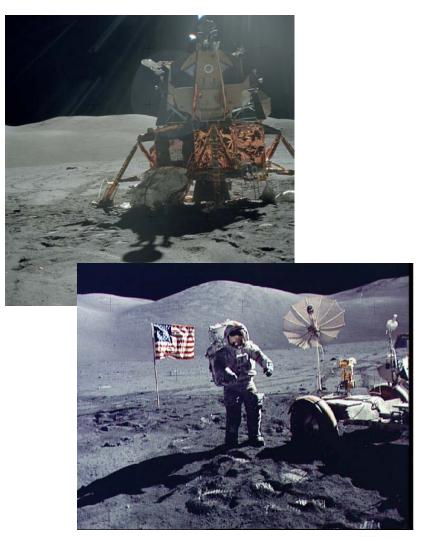
## **Topics**

- Historical use of Pu-238
- Projected future demand
- Current method
- Alternative method
- Preliminary results
- Summary



### Historical Use of Pu-238

- Pu-238 has been used in most space missions since the early days of Apollo
- RTGs still function on the lunar surface
- RTGs are on the farthest man-made object, Voyagers 1 and 2, now near 100 AU from Earth
- RHUs are on the rovers on Mars
- Domestic Production Ceased in 1988

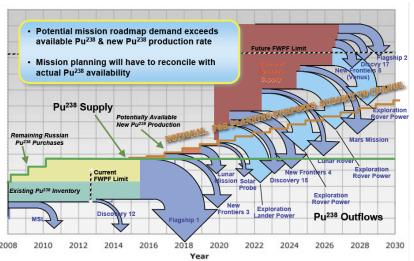




## NRC report

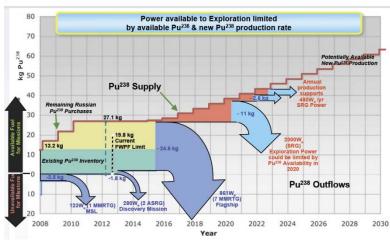
- Recently, NASA sponsored a National Research Council to convene a committee to review the status of Pu-238 production. Their final report, "Radioisotope Power Systems: An Imperative for Maintaining U.S. Leadership in Space Exploration" stated:
- "Plutonium-238 does not occur in nature. Unlike 239Pu, it is unsuitable for use in nuclear weapons. Plutonium-238 has been produced in quantity only for the purpose of fueling RPSs. In the past, the United States had an adequate supply of 238Pu, which was produced in facilities that existed to support the U.S. nuclear weapons program. The problem is that no 238Pu has been produced in the United States since the Department of Energy (DOE) shut down those facilities in the late 1980s. Since then, the U.S. space program has had to rely on the inventory of 238Pu that existed at that time, supplemented by the purchase of 238Pu from Russia. However, Russian facilities to produce 238Pu were also shut down many years ago, and the DOE will soon take delivery of its last shipment of 238Pu from Russia. The committee does not believe that there is any additional 238Pu (or any operational 238Pu production facilities) available anywhere in the world. The total amount of 238Pu available for NASA is fixed, and essentially all of it is already dedicated to support several pending missions-the Mars Science Laboratory, Discovery 12, the Outer Planets Flagship 1 (OPF 1), and (perhaps) a small number of additional missions with a very small demand for 238Pu. If the status quo persists, the United States will not be able to provide RPSs to ACE any subsequent missions." NUCLEAR

#### Demand for Pu-238 NASA Planning



NASA mission plans assuming a 1.5 kg/yr production rate of Pu-238- circa 2011.

Missions to outer planets planned by NASA circa 2010

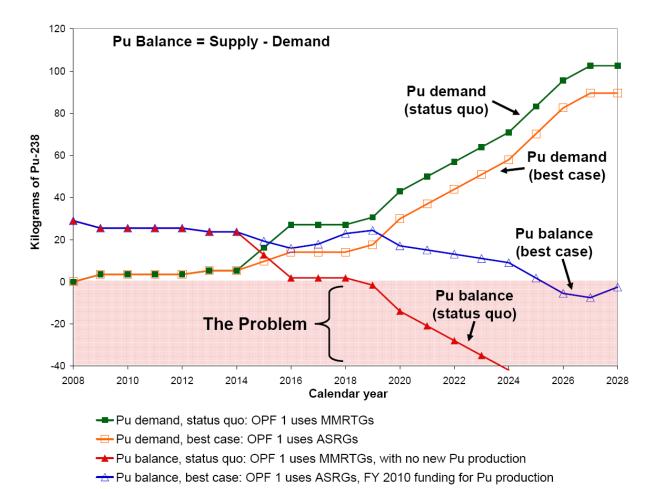


Radioisotope Power Systems: An Imperative for Maintaining U.S. Leadership in Space Exploration, National Research Council committee report. ISBN: 0-309-13858-2, 74 pages, (2009)



#### Projected Pu-238 balance in U.S. stockpile.

Note that the figure assumes production of 5kg/yr whereas current estimates are for a maximum of ~1.5kg/yr



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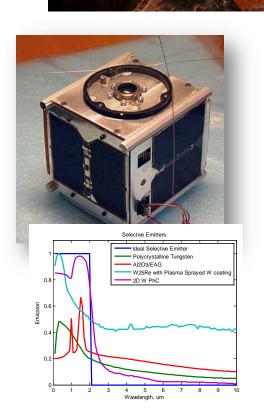
# The CSNR is developing future systems that require Pu-238

#### Mars Hopper

 Hop 6-10 km every 7 days for years – needs 2.5 kg Pu-238

#### Radioisotope Thermal Photo-Voltaic (RTPV)

- Small satellites are increasing in use in near Earth space
  - "Projections indicate a strong increase in nano/microsatellite launch demand, with an estimated range of 100 to 142 nano/microsatellites (1-50 kg) that will need launches globally in 2020 (versus 23 in 2011)."
    - -- Space Works Commercial report November 2011
- The use of micro or nano satellites offers the potential for cheaper exploration of the solar system
- The smallest nuclear source available will be the ASRG at 140 w with a mass of 22 kg
- No power source exists below the 100 w level to support small sat exploration beyond Mars orbit
- Pursuing RTPV development with NASA Ames offers potential for 50-70 kg/kw (X2 reduction in mass versus ASRG; 6X reduction versus MMRTG)
- Small sats could be applicable to deep space missions if a low–mass, radioisotope power supply is developed





## Production mechanism

Np-237 + n → Np-238 (β decay 2.1 d)→ Pu-238

- Losses
  - Np-237 + n → fission (.5 MeV threshhold)
  - Np-238 + n → fission (large cross section)
  - Np-237 (n,2n) Np236 contaminant
  - Pu-238 + n  $\rightarrow$  fission
- Implies short exposure in high flux and then removal for decay to Pu

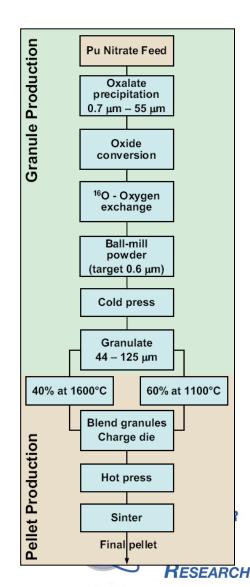


# Issues with current method

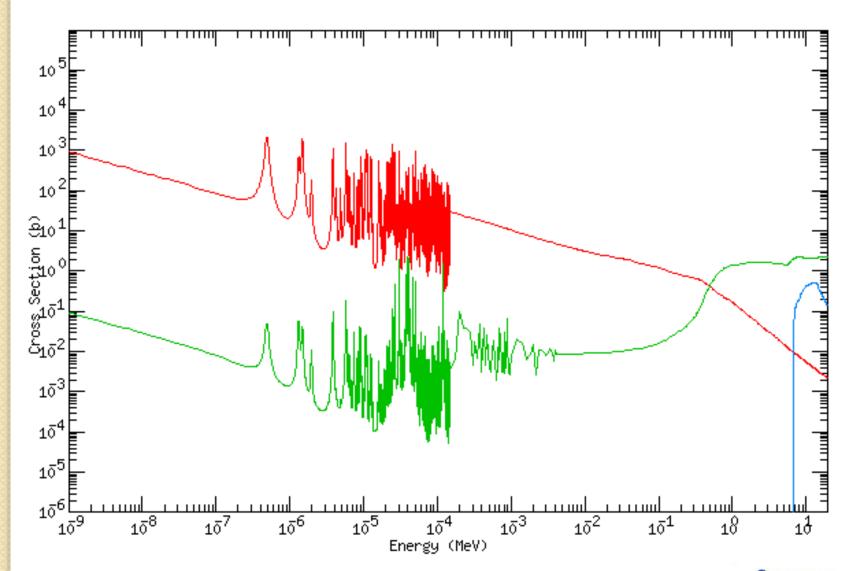
- Large mass of Np-237 is inserted into ATR or HFIR for long periods
  - Aluminum pins filled with NpO2
  - Irradiated for 6 mo to 1 yr
- Np-238 has a very large thermal neutron fission cross section – roughly 85% of the Np-238 created is fissioned
- Long irradiation creates a large inventory of fission products
  - Requires dissolving large, radioactive masses in acidic solution
  - Requires a large facility to handle the mass and the high radioactivity levels

#### Fabrication issues

- Ball milling of sub-micron powders leads to exposures
- Reconstitution of NpO2 from solution involves handling
- Costly and inefficient

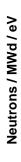


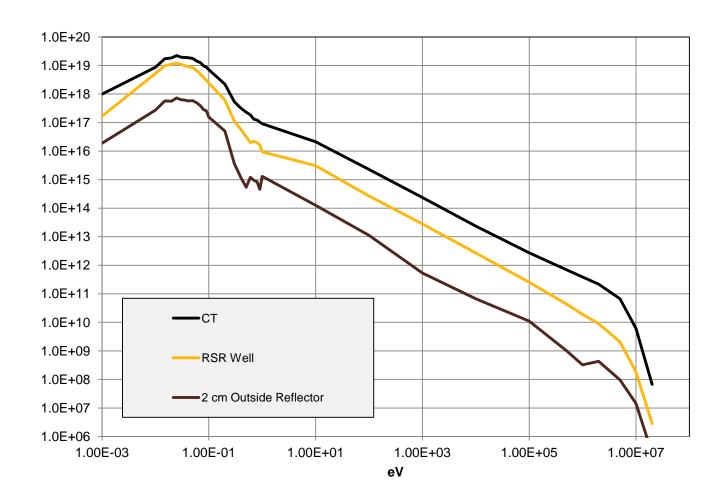
## Plot of the energy dependent microscopic cross section for <sup>237</sup>Np absorption in red, <sup>237</sup>Np fission in green and <sup>237</sup>Np to <sup>236</sup>Np in blue





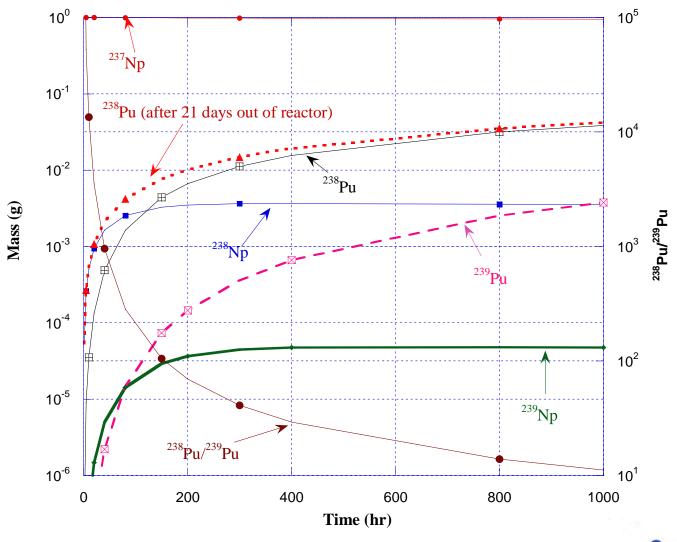
#### Neutron spectra at the 1 MW TRIGA at Kansas State University







#### **Production**



Isotope levels versus irradiation time for 1 g of Np-237 in a flux of 1e14 n/cm2-s (courtesy of Dr. Ken Czerwinski, UNLV)



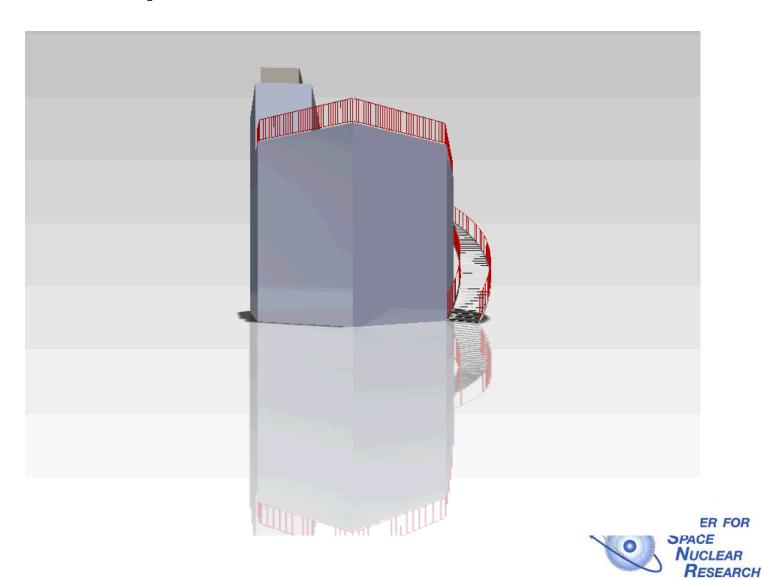
## Basics of Alternative Approach

- Slightly alter the configuration of a large, e.g. 5
  MW, licensed TRIGA to accommodate a loop around the core
- Continuously flow target material around the core
  - Residence time in the flux to be few days
- Allow Np-238 to decay for 5-10 half lives (up to 21 days) en route to processing facility.
- Separate Pu from other components in small, quantized batches using resin columns and established methods
- Re-inject run-off back into feed stream
- Allows small, university scale laboratory for processing facility- i.e. substantially reduced cost.



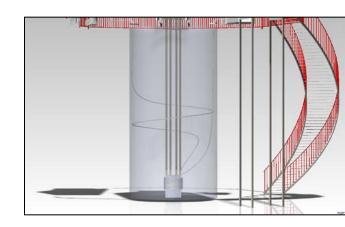


# Concept



# Continuous Target

- Allows short residence times and longer decay times
- Reduces fission product inventory and radioactivity levels
- Allows smaller processing lines
- Smaller facility





## Benefits

- Significantly simplifies or eliminates target fabrication and target processing facilities
- Reduces time to material production
- Make more efficient use of Np stockpile (less fission losses)
- Provides ability to tailor Pu-238 quality
- More economical operations
- Allows for production of other radioisotopes for medical and industrial use (duo use mode / shared investment)
- Does not require government capital construction funding (commercialization option)
- Government only pays for product received (commercialization option)



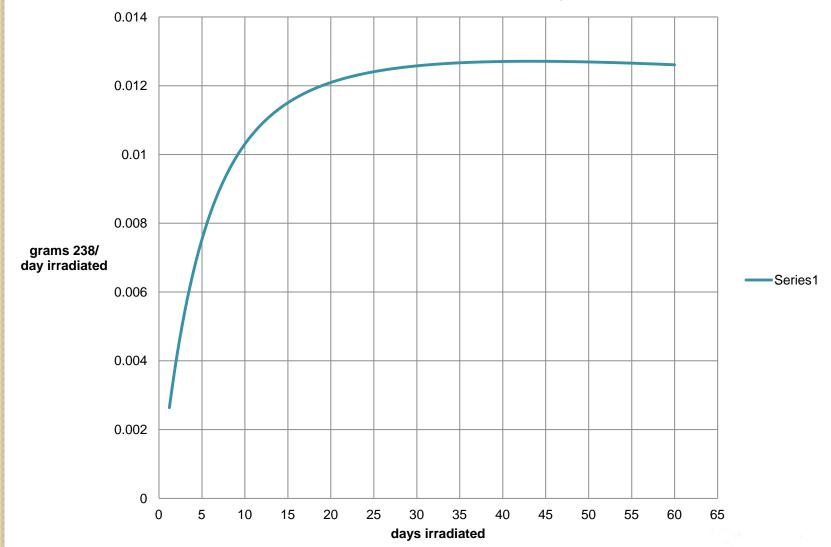
### Issues to answer

- Production
  - Impact on reactor operations from large amount of Np solution around core
  - Maximum concentration of Np possible and temperature dependence
  - Neutron spectral shift effect?
  - Residence and decay times for optimization
    - Fission product inventory time dependence and level versus amount of Pu-238 produced
  - Mechanical movement of hundreds of capsules
- Fabrication
  - Direct fabrication of PuO2 spheres from product solution
  - Fabrication of pellet with correct porosity and density profiles
- Political
  - US government must own all SNM. How will price be determined?
  - Use of DOE sites if chosen?

# Preliminary results



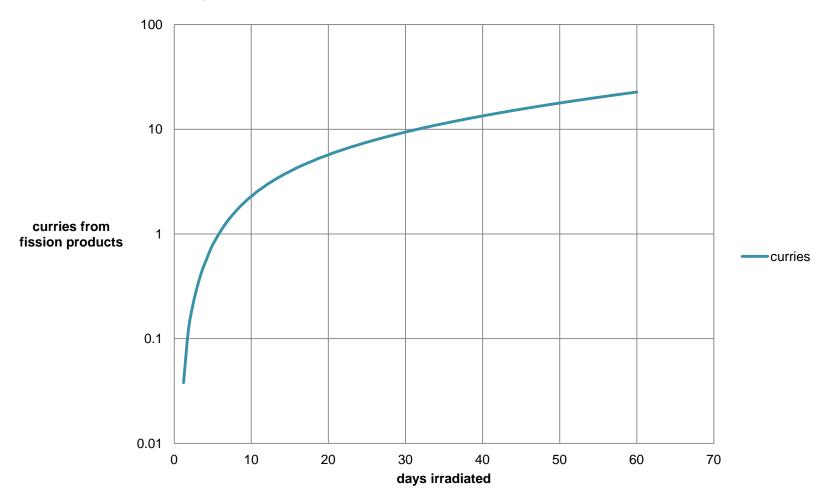
## 238Pu grams produced/gm Np237-irradiation day (1E13 flux, max is at 43.25days )





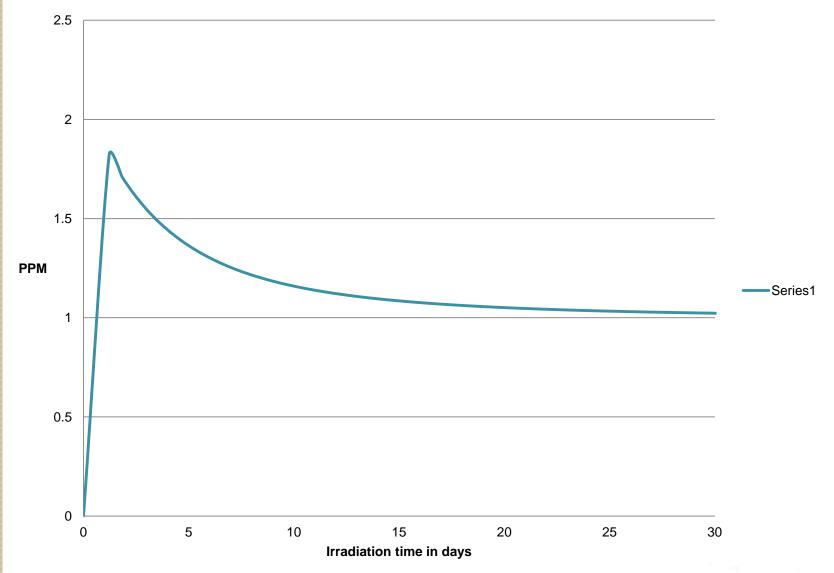
#### Curries per day of irradiation time

Assuming 6% 137Cs and 6% 99Tc based of fission yields





#### PPM 236Pu to 238Pu





### Planned POC experiments

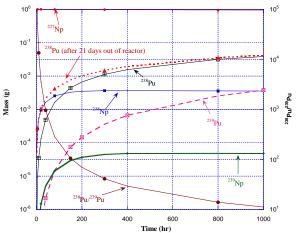
#### Irradiation to verify interaction rates

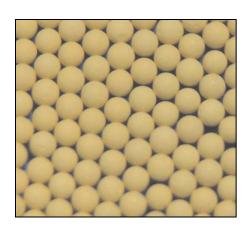
- •Difficult to calculate due to resonance region of cross section
- Verify production rate versus irradiation time
- Verify Pu-236 ratio and fission rate
- 4 day irradiation in the Kansas State Univ 1 MW reactor

# •Fabrication of PuO2 spheres for improved safety in handling

Direct route from nitrate solution to microsphere - (dust-free) with fewer steps

- Univ of Michigan fabricating spheres
  - CeO2 completed
  - •DUO2 scheduled as a surrogate for PuO2





Air-dried  $UO_3 \cdot 2H_2O$  microspheres with diameters of 1000 µm [9]



## Cost assessment

- Cost assessment is underway
- Assumes a private entity buys the reactor and sites it at
  - a) green field location or
  - b) DOE site.
- Has to trade security and handling costs versus transportation of Pu costs
- Will trade cost versus reactor power level
- Will determine the price charged to the government
- Preliminary results show that a 20% return can be met with a price of \$6M/kg

# Benefits of Continuous process

- Current process
  - Target material is NpO2-20 vol% - reconstituted after separation
  - Fuel clad interactions
  - Fission gas generation
  - 1000s gal of radioactive acidic waste per year\*
  - 10s of 5 gal drums of trans-uranic waste per year\*
  - Operating costs of HFIR and ATR are high

#### **Alternative Process**

- •Target material is solution that is compatible with separation process
- No cladding
- Fission is minimized
- Waste is estimated at gms/yr – nitric acid solution is recycled
- Reduced Pu236 content
- Operating costs of private small reactor are greatly reduced

# Alternative Option Conclusions

- Option for continuous target production of Pu-238 appears to be a viable, cost effective alternative
- Allows production quantities to be made in incremental stages- many kgs/yr
- Continuous production process allows small process footprint and minimal materials inventory
- Reduces government up front costs
- Places costs within reach of commercial venture ention